Generation of Terahertz Electromagnetic Pulses from Quantum-Well Structures

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Abstract—Terahertz generation from semiconductor quantum-well structures pumped by a femtosecond optical pulse is studied. We propose a three-level model for the electrons and holes in the quantum wells. We then solve the coupled optical Bloch equations directly using a Runge–Kutta method and calculate the terahertz radiation field. We study optical rectification and quantum beats caused by charge oscillations in 1) a coupled quantum well in which quantum beats occur between two electron states of the coupled system and 2) a single-quantum-well structure in which quantum beats occur between light-hole and heavy-hole excitons. Our theoretical results agree very well with the experimentally measured terahertz data.

I. INTRODUCTION

There has been a substantial research interest in the nonlinear optical properties of semiconductor quantum wells [1], [2]. The research is not merely of academic interest but is also significant for technological innovations. Modern technology has pushed the performance of optoelectronic devices well into the gigahertz regime; however, devices operating at the terahertz (THz) range will require the invention of novel structures and possibly involve very different physical processes. The basic functions of information systems would include generation, propagation, processing, and detection of femtosecond electrical signals. Much of the above has been accomplished in the past few years. The detection of an ultrashort electromagnetic pulse using semiconductor devices is usually based on a technique pioneered by Auston et al. [3], which utilizes photoconductive switch of the created carriers. This dipole antenna detection method [3], [4] can measure not only the amplitude but also the phase of the submillimeter-wave radiation field.

A scheme [5], [6] for femtosecond pulse generation by virtual excitations of semiconductor quantum wells was proposed in 1987. As shown in Fig. 1(a), the laser pulse has an energy $h\omega_{op}$ below the fundamental bandgap and does not generate any real carrier population, but it would induce an instantaneous polarization. Because no real carriers are involved in this process, an instantaneous electrical pulse as short as the optical pulse (~100 fs) is expected. Detection of this signal is complicated by the occurrence of two-photon absorption, which leads to real carrier excitation [7] and excitation in the Franz–Keldysh tail. Different methods have been proposed by several research groups. However, effects due to virtual excitations have never been observed, since the signal is expected to reduce significantly as the optical pump energy is detuned below the band edge. Although subbandgap excitation, or virtual excitation, of bulk GaAs has also been claimed to induce THz radiation [8], this result has yet to be confirmed by other groups.

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On the other hand, it has been shown recently that a THz electromagnetic pulse can be generated from the surface of many semiconductors [9], such as GaAs and InP, when the surface is illuminated with a femtosecond laser pulse with an optical energy \( h\omega_{\text{opt}} \) above the bandgap. The effect was initially attributed to the acceleration of photogenerated carriers across the surface depletion region. The resulting transient current \( J(t) \) would radiate an electromagnetic pulse \( E(t) \propto \frac{\partial P(t)}{\partial t} \), which has frequency components up to several terahertz. This model, although it was able to explain some of the observed phenomena, cannot explain an important observation, namely the variation of the field amplitude with the semiconductor crystallographic orientation. According to the current surge model, the radiated signal would be isotropic because the photoconductivity of a zinc-blende semiconductor is well-known to be isotropic; therefore, it should not vary with the crystal orientation. But the experimental data showed a twofold symmetry for (100) surfaces and a threefold symmetry for (111) surface. A new model [10] based on optical rectification involving the second-order nonlinear susceptibility of the material has recently been proposed. This model explains not only the crystal orientational dependence but also all of the other major features such as the polarity dependence on substrate dopants, the voltage bias dependence, and the temperature dependence. Its basic mechanism can be easily understood as shown in Fig. 1(b). Because of the depletion field, the centroids of the electron and hole wavefunctions are separated. The ultrashort laser pulse creates electron–hole pairs near the surface. Once generated, the electrons and holes are separated instantaneously and a net dc polarization density \( P(t) \) exists within the region. The transient polarization gives rise to a radiation field \( E_R(t) \propto \frac{\partial^2 P(t)}{\partial t^2} \). This mechanism is similar to the model for virtual excitations in quantum wells [5], [6], except that real carriers are being created. However, its magnitude has been shown [9] to be much stronger than that induced by the virtual excitation.

Recently, the observation of charge oscillations in coupled quantum wells by THz radiation measurements has been reported [11]. The laser excitation for those experiments is above the absorption band edge. In the past, quantum beats have been shown in degenerate four-wave mixing and time-resolved luminescence spectroscopy [12], [13]. However, the THz radiation experiment provides the first direct observation of the coherent oscillatory charge phenomenon associated with the quantum beats. It was found that an instantaneous THz signal is created by the incident laser pulse, followed by a ringing signal due to the charge oscillations in the coupled quantum wells. Since the quantum well limits the transport of created carriers perpendicular to the wells, the dominant mechanism can only come from the optical rectification effect. Similar results were obtained for a specially designed single-quantum-well structure [14], where terahertz radiation due to quantum beats between light-hole and heavy-hole excitations were observed for the first time.

In this paper, we model the terahertz generation due to both optical rectification and quantum beats in the time domain. The theoretical formulation is shown in Section II. Many-body effects are neglected since the photoexcited carrier density is very small (less than \( 5 \times 10^9 \text{ cm}^{-2} \)) in the experiments.

The exciton effects, which slightly affect the terahertz radiation frequency, are calculated using an exciton Green’s function method [15]. Numerical details of simulations for both the coupled-well and single-well structures are presented in Section III with comparisons with the experimental data.

The importance of the physical parameters, such as the laser bandwidth and dephasing time, will be discussed in Section IV. We then conclude in Section V.

II. THEORETICAL FORMULATION

The density-matrix approach works very well in the study of optical properties of electronic systems and will be used for this investigation. For the study of the optical rectification effect, a simple two-level model, as in Fig. 1(a), is sufficient. A more sophisticated three-level model is required to explain the quantum beats. In a more general sense, both optical rectification and quantum beats can all be described in nonlinear optics theory by the difference–frequency mixing within the bandwidth of the laser pulse. The generated terahertz signal is just a manifestation of the coherent interaction of the photon with various electron–hole exciton states. A schematic diagram for the three-level model is shown in Fig. 2(a), which has two conduction-band levels for electrons and one valence-band state for heavy or light holes. We start with the basic Liouville equation

\[
\frac{i\hbar}{\partial t}\rho = \{H, \rho\} = [H_0 - \mu \cdot E, \rho] + i\hbar \left( \frac{\partial \rho}{\partial t} \right)_{\text{relax}}
\]

where \( \rho \) is the density matrix operator, \( H \) is the total Hamiltonian, and \( H_0 \) is the unperturbed Hamiltonian. In (1), the dipole approximation is used for the description of the perturbing laser field with \( \mu \) the dipole moment and \( E \) the optical electric field. The last term in (1) accounts for the recombination and
scattering relaxations. Because the duration of the laser pulse is about one hundred femtoseconds and its amplitude is smooth within an optical cycle, the laser field amplitude is assumed to be

\[ E(t) = E_p(t)e^{-i\omega_{op}t} + E_p^{*}(t)e^{i\omega_{op}t} \]  

(2)

where the envelope function of the pump field \( E_p(t) \) is slowly varying compared with the optical frequency \( \omega_{op} \). We define the density-matrix elements, keeping in mind that the optical energy is close to the bandgap of the semiconductor. Therefore, the rotating wave approximation is used, which assumes that all but the resonant terms are negligible. Let

\[ \rho_{11} = \rho_{11}(t), \rho_{22} = \rho_{22}(t), \rho_{33} = \rho_{33}(t), \rho_{12} = \rho_{12}(t), \rho_{13} = \rho_{13}(t)e^{-i\omega_{op}t}, \rho_{23} = \rho_{23}(t)e^{-i\omega_{op}t} \]  

(3)

where \( \rho_{nm} = <n|\rho|m> \) is the matrix element of the original density operator; \( \rho_{nm} \) describes the mixing probability between energy states \( n \) and \( m \), and is equal to the population of the level if \( n = m \). The equations of motion for the density-matrix elements can be written explicitly as follows:

\[ \frac{\partial \rho_{11}(t)}{\partial t} = -\frac{\rho_{11}(t) - \rho_{11}^{(0)}}{T_1} + \frac{i}{\hbar}[\mu_{13}\rho_{31}(t)E_p(t) - \mu_{31}\rho_{13}(t)E_p^{*}(t)] \]  

(4)

\[ \frac{\partial \rho_{22}(t)}{\partial t} = -\frac{\rho_{22}(t) - \rho_{22}^{(0)}}{T_2} + \frac{i}{\hbar}[\mu_{23}\rho_{32}(t)E_p(t) - \mu_{32}\rho_{23}(t)E_p^{*}(t)] \]  

(5)

\[ \frac{\partial \rho_{33}(t)}{\partial t} = -\frac{\rho_{33}(t) - \rho_{33}^{(0)}}{T_3} - \frac{i}{\hbar}[\mu_{13}\rho_{31}(t) + \mu_{23}\rho_{23}(t)]E_p^{*}(t) \]  

(6)

\[ \frac{\partial \rho_{12}(t)}{\partial t} = -\frac{i(\omega_{12} + \frac{1}{T_{12}})\rho_{12}(t)}{\hbar} + \frac{i}{\hbar}[\mu_{13}\rho_{13}(t) + \mu_{32}\rho_{32}(t)]E_p^{*}(t) \]  

(7)

\[ \frac{\partial \rho_{13}(t)}{\partial t} = -\frac{i(\Delta_{13} + \frac{1}{T_{13}})\rho_{13}(t)}{\hbar} + \frac{i}{\hbar}[\mu_{13}\rho_{33}(t) - \mu_{31}\rho_{13}(t)]E_p^{*}(t) \]  

(8)

\[ \frac{\partial \rho_{23}(t)}{\partial t} = -\frac{i(\Delta_{23} + \frac{1}{T_{23}})\rho_{23}(t)}{\hbar} + \frac{i}{\hbar}[\mu_{23}\rho_{32}(t) - \mu_{32}\rho_{23}(t)]E_p^{*}(t) \]  

(9)

where we define the detuning energies \( \hbar\Delta_{13} = \epsilon_{13} - \omega_{op} \) and \( \hbar\Delta_{23} = \epsilon_{23} - \omega_{op} \), and the energy level splitting \( \hbar\omega_{12} = \omega_{12} = \epsilon_{23} - \epsilon_{13} \). The interband transition energy \( \epsilon_{nm} \) including the \( k \)-dependence is defined as

\[ \epsilon_{nm} = \epsilon_{n} + \epsilon_{m} + E_g + \frac{\hbar^2k^2}{2m_r^*} \]  

(10)

\[ \frac{1}{m_r^*} = \frac{1}{m_c} + \frac{1}{m_v}. \]  

(11)

Here \( \epsilon_n \) is the \( n \)th conduction subband-edge energy, \( \epsilon_m \) is the \( m \)th valence subbed-edge energy, \( E_g \) is the bandgap of bulk material, and \( m_r^* \) is the reduced effective mass. The interband-dipole-matrix elements for heavy holes under the parabolic band model are [15]

\[ \mu_{13} = \frac{eP}{\epsilon_{13}} \langle \phi_1(z)|\phi_3(z)\rangle \sqrt{\frac{1}{4}(1 + \cos^2 \theta_{13})} \]  

(12)

\[ \mu_{23} = \frac{eP}{\epsilon_{23}} \langle \phi_2(z)|\phi_3(z)\rangle \sqrt{\frac{1}{4}(1 + \cos^2 \theta_{23})} \]  

(13)

\[ P = \frac{\hbar^2cE_g}{2m_c \left( \frac{E_g + \frac{2}{3}\Delta}{} \right)} \]  

(14)

\[ \cos \theta_{nm} = \frac{\epsilon_n + \epsilon_m}{\epsilon_n + \epsilon_m + \frac{\hbar^2k^2}{2m_r^*}} \]  

(15)

The intersubband dipole-moment elements are defined as

\[ \mu_{nm} = -|e|\langle \phi_n(z)|\phi_m(z)\rangle = -|e|\epsilon_{nm}. \]  

(16)

The relaxation term in (1) is represented by the phenomenological decay time constants, i.e., the longitudinal decay times \( T_{11}, T_{22}, \) and \( T_{33} \), and the dephasing times \( T_{12}, T_{13}, \) and \( T_{23} \). Note that the trace of the density matrix elements satisfies the conservation

\[ \rho_{11}(t) + \rho_{22}(t) + \rho_{33}(t) = 1 \]  

(17)

if we assume the longitudinal times are the same, \( T_{11} = T_{22} = T_{33} \) and sum up (4)-(6). We solve the nonlinear optical Bloch equations (4)-(9) by the 5th-order Runge-Kutta method, which is very efficient for solving ordinary differential equations if all of the characteristic time constants are of the same order. In our two-band (three-level) model, the density-matrix elements at different \( k \) values are decoupled since the exciton effects are ignored. It is noted that many-body effects [16] [17] have been ignored in our formulation since the photoexcited carrier density is less than \( 5 \times 10^{19} \text{cm}^{-2} \). This simplifies our numerical procedure by a tremendous amount, yet, as we will show later in our numerical results, this simplified theory explains most features of the experimental data. A more elaborate calculation [18] taking into account many-body effects shows no substantial difference from the results obtained using these simplified equations (4)-(9) except for the energy shift due to the exciton effects, which will be calculated using an exciton Green's function approach. Similarly in transient four-wave-mixing experiments, it has been pointed
out [19] that many-body effects do not substantially alter the temporal positions of constructive and destructive interference in a quantum beat experiment, although they may affect the rise and decay times.

Once all the density-matrix elements at each $k$ value are found, we perform an integration over $k$-space to obtain the induced dc polarization density

$$P(t) = \frac{|e|}{V} \sum_k [z_{33} - z_{11}] \rho_{11}(t) + (z_{33} - z_{22}) \rho_{22}(t) - 2z_{12} \text{Re} (\rho_{12}(t)). \quad (18)$$

The laser pulse shape can be either a Gaussian or sech function. In our calculations, we assume the light pulse field to be sech $(t/\tau)$ with a pulse duration of 1.763 $\tau$, full-width at half-maximum (FWHM). Similar equations can be obtained with modifications of the energies and the dipole matrix elements for a three-level system as shown in Fig. 2(b), which has two valence-band states and one electronic state.

III. CHARGE OSCILLATIONS IN QUANTUM WELLS

From quantum mechanics, we know that in a W-shaped potential system with two energy eigenstates, a particle, initially created within one well, will oscillate back and forth between the two potential traps. This is true only if the two levels remain coherently coupled. This model has been successfully applied to the explanation of the ammonia molecular vibration, chemical reactions, and the interaction of the system with a dissipative reservoir. In semiconductors, optical transitions are often inhomogeneously broadened, making the observation of this oscillatory phenomenon difficult. It was made possible only recently due to the advances in crystal growth and the availability of stable femtosecond optical pulses. The dephasing time of free carriers is very short, about 100 fs, but the formation of excitons in bulk GaAs and quantum wells may have a dephasing time up to a few picoseconds at low temperatures [20]. A degenerate four-wave-mixing (FWM) measurement [21] showed that if the excitation pulse has a photon energy centered at the exciton energy, the FWM signal is dominated by excitons. The long dephasing times of excitons combined with a good interfacial crystal quality leads to the successful observation of charge oscillations [11]. In this section, we will discuss two different quantum-well structures. For an asymmetric coupled-well design, the potential profile is similar to the W-shaped system, and the excitons in either the narrow well or the wide well can be selectively pumped. The second system has only a single quantum-well structure but still has the oscillatory THz signals caused by the quantum beats between the light-hole and heavy-hole excitons due to the valence subband energy dispersion relations in a quantum well.

IV. COUPLED QUANTUM-WELL STRUCTURE

The sample used in [11] consists of ten pairs of a wide-well (WW) and a narrow-well (NW) region separated by a 25-Å-thick $\text{Alo}_{0.2}\text{Ga}_{0.8}\text{As}$ barrier. Both wells are made of a GaAs layer, which is 145 Å for WW and 100 Å for NW.
Fig. 4. Experimentally measured terahertz electromagnetic radiations from the coupled quantum wells (after [11]) at five bias fields. The resonant field is near 10.5 kV/cm.

Fig. 3(c), the lowest eigenstate is in the narrow well, and the coupling between the two electronic states will decrease with increasing field. We have calculated the eigenstate wavefunctions for the three bias conditions and the results are shown in Figs. 5(a)–(c). It can be seen that the switching of the lowest state (the magnitude squared of the wavefunction is in solid curves) from the wide well, Fig. 5(a), to the narrow well, Fig. 5(c), is evident. The interband bandedge transition energies are shown in Fig. 6 as a function of the bias field. The Coulomb interactions between the electrons and holes are not included in Fig. 6, but they will be taken into account later. The transition energies as a function of electric field for the heavy hole in the wide well to the two electronic states, designated as hh–WW, show a pair of hyperbolic curves (solid lines) with a minimum splitting at the resonant field. This is the “anticrossing” of the two interband excitations. For the transitions from the heavy hole in the narrow well, there is an anticrossing at the same bias, but the energy curves (dashed lines) generally show an increase with an increasing field because of the bias dependence of the NW heavy-hole subband energy.

With the above information, the induced dc polarization and the terahertz radiation field can be calculated. The results are as shown in Figs. 7 and 8. Figure 8 should be compared with the experimental results in Fig. 4. The initial response of the system shows an adiabatic following of the pump pulse because the densities of the generated electron–hole pairs follow the light pulse shape. According to (18), the total polarization density is proportional to the net separation between the electron and hole, \((z_{33} - \rho_1)\) and \((z_{33} - \rho_2)\), multiplied by the carrier population in level 1 and level 2, \(\rho_1\) and \(\rho_2\), respectively. The generated carrier density is proportional to the laser intensity and interband dipole moments. To see how the radiation intensity changes with the bias, the dipole moments are plotted in Fig. 9 versus the dc field. The interband dipole moments are proportional to the overlap of electron and hole wavefunctions; hence, the two solid curves in Fig. 9(a) switch at the resonant field. The net displacement of the dipole moment in Fig. 9(b) shows the same tendency. The combined result of this field dependence is that the instantaneous signal amplitude, in Fig. 8, initially increases with the bias field, reaches its peak at the resonance field, but then decreases at a higher field. This agrees with the experimental observation in Fig. 4. The induced polarization \(P(t)\) in Fig. 7 begins to show the oscillating feature after the 1-ps period. The oscillation frequency corresponds to the energy splitting of the two electronic states and is strongly field-dependent, as discussed in the previous paragraph. This is different from the
Fig. 7. Theoretical results of the induced dc polarization density at several bias conditions. The dephasing time $T_{12}$ is assumed to be 1.2 ps for 1.4 kV/cm, 2 ps for 6.7 kV/cm, 1.3 ps for 10.5 kV/cm, 0.7 ps for 13.2 kV/cm, and 0.4 ps for 15.8 kV/cm.

Experimental observation, which shows very little dependence on the applied bias, and the oscillation frequency is 1.5 THz. The difference can be explained if we take into account the electron–hole Coulomb interaction, i.e., the excitonic effect.

Excitonic effects result in several modifications of the energy-band structures. We have calculated the exciton energies for the coupled quantum-well structure using an exciton Green's function method [15]. The advantage of this approach is that the Green's function takes into account the coupling between multibands and gives a more accurate estimate of the oscillator strength. We calculate the electron and hole subband energies with corresponding wave functions first, then solve the Schrödinger equation in the momentum space taking into account the electron–hole Coulomb interaction. The linear optical absorption spectrum is then calculated to extract the exciton energies. This method has been shown to give very good agreement with experimental data [15]. Our result for the coupled quantum wells is shown in Fig. 10 for the hh-WW and hh-NW excitons. Two major modifications are observed. 1) The binding energies of the interwell and intrawell excitons are different. Because of the large overlap of electron and hole wavefunctions for the intrawell excitons, they have a stronger Coulomb attraction and hence a larger binding energy than that of the interwell excitons. Comparing the bandedge energies in Fig. 6 and the exciton transition energies in Fig. 10, we find that the binding energy is about 9 meV for an intrawell exciton and 5 meV for an interwell exciton. 2) The resonant field is different for the NW and WW states. The resonant field for hh-NW excitons is at 4 kV/cm, which is smaller than the original value 8 kV/cm without the exciton effects. For hh-WW excitons, the resonant field changes to 10.5 kV/cm, which is close to the experimental value. The main reason is due to the difference between the heavy-hole state wavefunctions in the WW and in the NW [22].

B. Single Quantum Wells

In analogy to the previous discussions of coupled quantum-well structures, we expect to observe charge oscillations from...
holes in certain structures. In GaAs, the heavy-hole effective mass \( m^*_{\text{hh}} = 0.45 m_0 \) and light-hole effective mass \( m^*_{\text{lh}} = 0.085 m_0 \), where \( m_0 = 9.1095 \times 10^{-31} \text{ kg} \) is the free-electron mass. According to the parabolic band model, the crystal Bloch functions for holes are orthogonal to each other; thus, there is no light-hole and heavy-hole transition dipole moment, which is required for the emission of THz radiation. But the valence band structures of quantum wells calculated using the Luttinger–Kohn Hamiltonian indicate that there are strong couplings between these two types of holes. According to the valence-band-mixing model, we would expect quantum beats from the coherent excitations of \( \text{lh} \) and \( \text{hh} \) excitons, which are separated in energy. Recently, Planken et al. [14] have designed a special quantum-well sample, and their results show clearly the importance of valence-band mixing.

The sample consists of an intrinsic multiple-quantum-well (MQW) region that has 15 periods of 175-Å GaAs single wells separated by 150-Å \( \text{Al}_{0.3}\text{Ga}_{0.7}\text{As} \) barriers. The substrate is \( n^+ \)-doped GaAs with a strain-relief layer sandwiched between the substrate and the MQW region. A metal-in Schottky diode is made from this sample with 50 Å of semitransparent chromium deposited as the metal contact. The sample is mounted in a continuous-flow liquid helium cryostat and the temperature is maintained at 10 K. The experimental setup is similar to the one described before except that the laser pulse has a wider bandwidth to cover the desired range. The detected THz waveforms as a function of electric field are shown in Fig. 11. The general characteristics are the same as those discussed in Section IIIA. There is a clear evidence of quantum beats from the light-hole and heavy-hole intersubband transitions. But contrary to the coupled well, the oscillation frequency shows a strong field dependence, as indicated in the inset. A note on the measurement is that the photon energy has to be tuned to cover both the \( \text{lh} \) and \( \text{hh} \) excitons to see the oscillations. If the pump photon energy is tuned below the resonance region, only an instantaneous electrical transient is observed.

We will model the experimental results including the valence-band mixing effects. Consider a three-level model as shown in Fig. 2(b), which has a heavy hole as state 1, a light hole as state 2, and an electron as state 3 in a quantum-well structure. As indicated in (18), the oscillating signal is related to the momentum-matrix element [26] as

\[
\langle m|\hat{e}\cdot\mathbf{r}|m'\rangle = \sum_{\nu,\nu'} \frac{1}{\hbar} \langle m, \nu|\hat{e}\cdot\mathbf{r}|m', \nu'\rangle
\]

where

\[
\begin{align*}
P &= \frac{\hbar^2}{2m_0} \gamma_1 (k_x^2 + k_y^2 + k_z^2) \\
Q &= \frac{\hbar^2}{2m_0} \gamma_2 (k_x^2 + k_y^2 - 2k_z^2) \\
L &= -i\sqrt{3} \frac{\hbar^2}{m_0} \gamma_3 (k_x - ik_y)k_x \\
M &= \frac{\hbar^2}{2m_0} \sqrt{3} \gamma_2 (k_x^2 - k_y^2) - i\frac{\hbar^2}{m_0} \sqrt{3} \gamma_3 k_x k_y
\end{align*}
\]

and the basis functions are shown in (20) at the bottom of this page, where \( \nu = 3/2, 1/2, -1/2, -3/2 \). The intersubband dipole-matrix is related to the momentum-matrix element [26] as

\[
\langle m|\hat{e}\cdot\mathbf{r}|m'\rangle = \sum_{\nu,\nu'} \frac{1}{\hbar} \langle m, \nu|\hat{e}\cdot\mathbf{r}|m', \nu'\rangle
\]

where

\[
\begin{align*}
3/2, 3/2 &= \frac{1}{\sqrt{3}} |X + iY| > \\
3/2, 1/2 &= \frac{1}{\sqrt{6}} (X + iY) |+2Z| > \\
3/2, -1/2 &= \frac{1}{\sqrt{6}} (X + iY) |+2Z| > \\
3/2, -3/2 &= \frac{1}{\sqrt{3}} (X + iY) |>
\end{align*}
\]
and can be expressed in terms of the envelope functions \( F_v(m, k_{ll}, k_z) \) [25]:

\[
\frac{\hbar}{m_0} <m, \nu|\hat{\epsilon}|p|m', \nu'> = \sum_{k_z} F^*_v(m, k_{ll}, k_z) F_{v'}(m', k_{ll}, k_z)
\]

\[
\times \left[ \sum_i e_i k_{ll} [2 \delta_{ij} D_{v'v}^{ij} + (1 - \delta_{ij}) D_{v'v}^{ij}] \right]
\]

\[
= \sum_i e_i \left[ \sum_{k_z} F^*_v(m, k_{ll}, k_z) F_{v'}(m', k_{ll}, k_z) \right]
\]

\[
\times [2 \delta_{kk} D_{v'v}^{k} + (1 - \delta_{kk}) D_{v'v}^{k}]
\]

\[
+ \sum_{k_z} e_i \left[ \sum_{k_z} F^*_v(m, k_{ll}, k_z) F_{v'}(m', k_{ll}, k_z) \right]
\]

\[
\times [k_{ll} (2 \delta_{kk} D_{v'v}'^{kk} + (1 - \delta_{kk}) D_{v'v}'^{kk})
\]

\[
+ k_z (2 \delta_{kk} D_{v'v}'^{kk} + (1 - \delta_{kk}) D_{v'v}'^{kk})]
\]

\[
= \hat{\epsilon} \cdot (P_{v'v} Q_{v,v'}^m + Q_{v,v'}^m S_{v,v'})
\]

(22)

where

\[
O_{v,v'}^{m,m'} = \int dz F^*_v(m, k_{ll}, z) F_{v'}(m', k_{ll}, z)
\]

(23)

\[
S_{v,v'}^{m,m'} = \int dz F^*_v(m, k_{ll}, z) \frac{\partial}{\partial z} F_{v'}(m', k_{ll}, z)
\]

(24)

Both the \( \hat{\epsilon} \cdot P_{v'v} \) and \( \hat{\epsilon} \cdot Q_{v,v'} \) matrix elements can be expressed in matrix form

\[
\frac{\hbar^2}{m_0} \begin{bmatrix} A_1 + A_2 & B & C \\
B^* & A_1 - A_2 & 0 & C \\
C^* & 0 & A_1 - A_2 & -B \\
0 & C^* & -B^* & A_1 + A_2 \end{bmatrix}
\]

(25)

where \( A_1, A_2, B, \) and \( C \) are tabulated in Tables I and II. The intersubband dipole moment between subbands \( m \) and \( m' \) is to sum over \( v \) and \( v' \) using (21)–(25) and Tables I and II. It can also be proven [26] that the same results for \( \hat{\epsilon} \) along \( z \) can be obtained from the following formulation.

\[
\mu_{\text{mm'}}^{\sigma}(k_{ll}) = -|\epsilon| \left[ \int dz g_m^{(1)}(k_{ll}, z) g_m^{(1)}(k_{ll}, z) + \int dz g_m^{(2)}(k_{ll}, z) s_m^{(2)}(k_{ll}, z) \right] \text{for } \sigma = U
\]

\[
= -|\epsilon| \left[ \int dz g_m^{(3)}(k_{ll}, z) g_m^{(3)}(k_{ll}, z) + \int dz g_m^{(4)}(k_{ll}, z) s_m^{(4)}(k_{ll}, z) \right] \text{for } \sigma = L
\]

(26)

where \( g_m^{(1)}(k_{ll}, z) \) and \( g_m^{(2)}(k_{ll}, z) \) are envelope functions associated with the upper Hamiltonian \( H^U \), and \( g_m^{(3)}(k_{ll}, z) \) and \( g_m^{(4)}(k_{ll}, z) \) are envelope functions associated with the lower Hamiltonian \( H^L \). The theoretical results from (21) and (26) are very close, and the latter is used for the simulation of induced radiation fields. Note the derivations of (22)–(26) show the importance of including the higher-order terms in the basis functions for the Bloch periodic parts under the effective-mass approximation.

The valence-band energy can be obtained by solving the block-diagonalized Luttinger–Kohn Hamiltonian. One example is shown in Fig. 12 for the flatband condition. At the zone center, the top curve (solid) corresponds to the hh state and the second curve (long-dashed) to \( \ell \) state. The two curves have a "crossing" behavior near \( k_{ll} = 0.01 \) Å\(^{-1} \), which is an indication of \( \ell \) and \( hh \) coupling due to the quantum confinement effects. The dipole moment \( \mu_{12} \) as a function of \( k_{ll} \) vector, where \( m = 1 = hh \) and \( m' = 2 = \ell \) in (26), is plotted in Fig. 13 for several bias fields. The regions with \( k_{ll} \) less than one inverse Bohr radius of excitons (about 0.01 Å\(^{-1} \)) give significant contributions. From these data, we can calculate the emitted terahertz waveforms by solving for the density-matrix elements for each \( k_{ll} \) using the optical Bloch equations (4)–(9), and the results for the radiation field by summing over all \( k_{ll} \) are shown in Fig. 14. For the 0 kV/cm curve, there exists an instantaneous signal (quantum beat) because of \( \mu_{12} \), which is nonzero at flatband, even though the self-dipole moments \( (211-233) \) and \( (222-233) \) are zero. Note that the expression (18) for the induced polarization density has to be multiplied by a minus sign here since levels 1 and

---

**TABLE I**

<table>
<thead>
<tr>
<th>( \hat{\epsilon} \cdot P )</th>
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<td>( \hat{\epsilon} \cdot \hat{P} )</td>
</tr>
<tr>
<td>( A_1 )</td>
<td>( \gamma_{1k_{ll}} )</td>
<td>( \gamma_{1k_{ll}} )</td>
</tr>
<tr>
<td>( A_2 )</td>
<td>( \gamma_{2k_{ll}} )</td>
<td>( \gamma_{2k_{ll}} )</td>
</tr>
<tr>
<td>B</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

**TABLE II**

<table>
<thead>
<tr>
<th>( \hat{\epsilon} \cdot Q )</th>
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<th>( \hat{\epsilon} \cdot \hat{Q} )</th>
</tr>
</thead>
<tbody>
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<td></td>
<td></td>
<td>( \hat{\epsilon} \cdot \hat{Q} )</td>
</tr>
<tr>
<td>( A_1 )</td>
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<td>0</td>
</tr>
<tr>
<td>( A_2 )</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>B</td>
<td>( -i \sqrt{3} \gamma_3 )</td>
<td>( -\sqrt{3} \gamma_3 )</td>
</tr>
<tr>
<td>C</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

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Fig. 12. The valence subband energies in the \( k \) space for zero bias.
Fig. 13. The intersubband (hh1 and lh1) dipole moments at four different bias fields.

Fig. 14. Theoretical results of the induced terahertz radiation caused by quantum beats between light holes and heavy holes.

2 now describe holes instead of electrons. In Fig. 15(a), the interband dipole moments $\mu_{13}$ (filled circles) and $\mu_{23}$ (open circles) tend to decrease with increasing field, which is due to the quantum-confinement Stark effect. The net displacements of the dipole moments (along the negative z direction) in Fig. 15(b) show a monotonic increase in magnitude with the field, but eventually saturate because both holes and electrons are pushed against the walls of the well. These results lead to the saturation of the instantaneous signal at high fields.

Fourier transform results of the charge oscillation region are shown in Fig. 16. The peaks of the frequency spectra correspond to the hh and hh excitations. Better results would be obtained if the Coulomb interactions are taken into account. In Fig. 17, we show the experimental data from 1) the peaks of the terahertz radiation (dots, see also inset in Fig. 11), 2) the energy splittings (crosses) between the lh and hh excitons, and with calculated results for the lh–hh energy splittings with (solid curve) and without (dashed curve) exciton effects. The energy splittings including the exciton effect are obviously closer to the experimental results (dots and crosses). Both the photocurrent spectra and the calculations with exciton effects for the lh–hh splittings also indicate that the terahertz radiation is generated by the quantum beats between the lh and hh excitons.

V. DISCUSSION

There are a few physical parameters that are very important in determining the emission of THz signals. Both the photon energy and bandwidth determine the possible interband excitations and the magnitude of the signal. One example is shown in Fig. 18 for the excitation of hh–ww excitons, whose energies are 1.5310 eV and 1.5352 eV, respectively. The bandwidth of the pump beam is assumed to be 7 meV.
electromagnetic transient comes from the adiabatic following of the laser pulse and is sensitive to the photon energy; the ringing of the THz oscillation is due to the quantum beats caused by charge transfer between a coupled quantum-well structure of the hh and lh intersubband transitions. The dephasing time and laser bandwidth are very important in determining the magnitude and shape of the oscillatory signal.

REFERENCES


Fig. 18. Calculated terahertz signals from a coupled quantum-well system with laser pulse energy ranging from 1.53 eV to 1.54 eV.

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\textbf{Martin C. Nuss}, photograph and biography not available at the time of publication.